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ABSTRACT

This report describes the technical progress made on the Pittsburgh Air Quality Study (PAQS) during the period of September 2004 through February 2005. Significant progress was made this project period on the analysis of ambient data, source apportionment, and deterministic modeling activities. The major experimental achievement this project period was the characterization of the mercury and fine particle emissions from two modern, large, commercial pulverized coal boilers. This testing completes the field work component of the Source Characterization Activity.

This report highlights results from mercury emission measurements made using a dilution sampler. The measurements clearly indicate that mercury is being transformed from an oxidized to an elemental state within the dilution. However, wall effects are significant making it difficult to determine whether or not these changes occur in the gas phase or due to some interaction with the sampler walls.

This report also presents results from an analysis that uses spherical aluminum silicate (SAS) particles as a marker for primary $PM_{2.5}$ emitted from coal combustion. Primary emissions from coal combustion contribute only a small fraction of the $PM_{2.5}$ mass (less than 1.5% in the summer and less than 3% in the winter) at the Pittsburgh site. Ambient SAS concentrations also appear to be reasonably spatially homogeneous. Finally, SAS emission factors measured at pilot-scale are consistent with measurements made at full-scale.

This report also presents results from applying the Unmix and PMF models to estimate the contribution of different sources to the $PM_{2.5}$ mass concentrations in Pittsburgh using aerosol composition information. Comparison of the two models shows similar source composition and contribution for five factors: crustal material, nitrate, an Fe, Mn, and Zn factor, specialty steel production, and a cadmium factor. PMF found several additional factors. Comparison between source contributions for the similar factors shows reasonable agreement between the two models. The sulfate factor shows the highest contribution to local $PM_{2.5}$ with an annual average contribution of approximately 28% (from PMF). The nitrate, crustal material, and primary OC and EC factors also show significant contributions on the order of 10-14%. The sulfate factor is affected by photochemistry and therefore shows maximum values in summer.

TABLE OF CONTENTS

Disclaimer	2
Abstract	3
Table of Contents	4
List of Figures	5
Excutive Summary	6
Experimental	7
Activity 1. Project Management	7
Activity 2. Ambient Monitoring	7
Activity 3. Source Characterization.....	7
Activity 4. Source Apportionment	7
Activity 5. Three-Dimensional Deterministic Modeling	8
Results and Discussion	8
1. Single Particle Characterization.....	8
4. Development and Evaluation of Measurement Methods.....	9
3. Effects of dilution on mercury speciation in coal-fired power plant plumes.....	10
4. Source Apportionment of Primary Coal Emissions with SAS.....	17
5. Source Apportionment using PMF and UNMIX	21
Conclusions	28
References	29
Presentations and Publications.....	29
Publications:.....	29
Presentations:	34

LIST OF FIGURES

Figure 1. Schematic of power plant. Mercury sampling with the dilution sampler was performed at the location labeled SCR Outlet.	10
Figure 2. Schematic of CMU Dynamic Dilution Sampler (Dilution Tunnel)	11
Figure 3. Time series of diluted and undiluted mercury concentration data taken on September 30	13
Figure 4. Flue gas and diluted sample mercury concentration. Note - Diluted samples have been corrected for tunnel blank and dilution ratio.	14
Figure 5. Ratios of diluted versus undiluted mercury concentrations. Diluted samples were blank and dilution corrected.	15
Figure 6. Time series of SAS number concentration and PM _{2.5} mass measured during the July 2001 intensive.	17
Figure 7. Time series of SAS number concentrations measured at the CMU site and the NETL site for January 2001.	18
Figure 8. SAS emission factors as a fraction of PM _{2.5} mass measured during pilot-scale and full-scale boiler testing.	20
Figure 9. Estimate PM _{2.5} mass contributed by primary coal emissions to the Pittsburgh Supersite during the Summer 2001 intensive.	21
Figure 10. Unmix source compositions apportioned by PM _{2.5} mass. All ten factors are outputs of PMF, while only the first six factors are outputs of Unmix. Although the Unmix regional transport factor includes primary OC and EC, it is graphed for comparison with the PMF sulfate factor.	23
Figure 11. PMF and Unmix source contributions apportioned by PM _{2.5} mass. All ten factors are outputs of PMF, while only the first six factors are outputs of Unmix. Note that for comparison purposes, the Unmix regional transport factor is compared to the PMF sulfate plus primary OC and EC factors.	24
Figure 12. Monthly average Unmix source contributions. Height of the bars corresponds to the monthly average PM _{2.5} mass measured with a TEOM. The study average represents the average source contributions from July 11, 2001 through July 31, 2002. Unmix uses PM _{2.5} mass as a fitting species so the mass of PM _{2.5} unexplained by Unmix is less than 1%	25
Figure 13. Monthly average PMF source contributions. Height of the bars corresponds to the monthly average PM _{2.5} mass measured with a TEOM. The study average represents the average source contributions from July 11, 2001 through July 31, 2002. The unexplained mass is the difference between the monthly average PM _{2.5} mass and the sum of the monthly averaged source contributions from each factor.	26
Figure 14. Linear regression results of PMF and Unmix source contributions.	27

EXECUTIVE SUMMARY

With support from the US Department of Energy and the US Environmental Protection Agency, Carnegie Mellon University is conducting detailed studies of the ambient particulate matter in the Pittsburgh, PA metropolitan area. The work includes ambient monitoring, source characterization, and modeling (statistical and deterministic) for source apportionment. The major objectives of the project include:

- To achieve advanced characterization of the PM in the Pittsburgh region. Measurements include the PM size, surface, volume, and mass distribution; chemical composition as a function of size and on a single particle basis; temporal and spatial variability.
- To obtain accurate current fingerprints of the major primary PM sources in the Pittsburgh region using traditional filter-based sampling and state-of-the-art techniques.
- To estimate the impact of the various sources (transportation, power plants, natural, etc.) on the PM concentrations in the area using both statistical and deterministic models.
- To quantify the responses of the PM characteristics to changes in these emissions in support of the emission control decision making in the area.
- To develop and evaluate current and next generation aerosol monitoring techniques for both regulatory applications and for determination of source-receptor relationships.

This document is the eighth semi-annual progress report for this project. During this project period significant progress was made on the analysis of ambient data, source apportionment, and deterministic modeling activities. Major achievements this project period and results described in this progress report include:

- Measurement of mercury and fine particle emissions from two full-scale pulverized coal boilers using a state-of-the-art dilution sampling system.
- Analysis of ambient and emission data for spherical aluminum silicate (SAS) particle concentrations to estimate the contribution of coal combustion to primary PM_{2.5} at the Pittsburgh site.
- Application of Unmix and PMF receptor models to apportion PM_{2.5} mass in the Pittsburgh area.
- Investigation of the effects of dilution on mercury oxidation state in coal-fired power plant plumes.

EXPERIMENTAL

This section provides an overview of the effort on various project activities. This project period the majority of the effort went into Activity 3 Source Characterization, Activity 4 Source Apportionment, and Activity 5 Three-Dimensional Modeling.

Activity 1. Project Management

During this project period additional data were submitted to EPA for inclusion in the Supersites Relational Database and to ATS for inclusion in the DOE-sponsored air quality database.

Activity 2. Ambient Monitoring

The purpose of this activity is to create an extensive database of ambient PM measurements for source apportionment, examination of aerosol processes, evaluation of instrumentation, and air quality model development and evaluation. Data collection is complete. Work has continued on the analysis of the ambient data set and selected results from this analysis are shown in the Results and Discussion section of this report.

Activity 3. Source Characterization

The purpose of this activity is to develop updated emission profiles for important source categories around Pittsburgh. Updated source profiles are being developed through a combination of source testing, fence line measurements, and analysis of highly time resolved data collected at the central site. These profiles are used in the source apportionment and deterministic modeling activities.

The major accomplishment in Activity 3 this project period was emission testing on two full scale coal-fired power plants. One test focused on effects of dilution on mercury partitioning on coal fired power plant plumes. The second test focused on measuring fine particle emission rates. The testing was done on modern, large (>400 MWe) pulverized coal fired boilers with low NO_x burners and electrostatic precipitators (ESP) for control of particle emissions. The mercury experiments were done on a boiler with an SCR unit for NO_x control. The particle characterization was performed downstream of the ESP. Neither plant had equipment for sulfur control. Both plants were operating on a blend of eastern bituminous and lower sulfur western sub-bituminous coal. This completes the field work component of the Activity 3. Selected results from the source characterization activities are shown in the Results and Discussion section of this report.

Activity 4. Source Apportionment

The purpose of this activity is to quantify the contribution of different sources to the fine PM_{2.5} levels in Pittsburgh. Significant effort was expended on the source

apportionment analysis this project period and selected results are shown in the Results and Discussion section.

Activity 5. Three-Dimensional Deterministic Modeling

The purpose of this activity is to evaluate the performance of the three-dimensional chemical transport model (PMCAMx) with air quality data collected by this and other projects. PMCAMx is a publicly available computer modeling system for the integrated assessment of photochemical and PM pollution. This CTM has been recently upgraded by the CMU team and ENVIRON to include state-of-the-art description of aerosol dynamics and thermodynamics, cloud chemistry, and wet removal processes. PMCAMx+ is the research version of the code and it includes the latest developments in Carnegie Mellon organic and inorganic aerosol and aqueous-phase chemistry modules. During this project period research focused on evaluating national inventories for primary organic carbon and elemental carbon used by the model.

RESULTS AND DISCUSSION

1. Single Particle Characterization

The University of California at Davis operated the RSMS-3, a single particle mass spectrometer, for twelve consecutive months (306 operation days) at the central monitoring site. This instrument provides semi-continuous, highly time resolved measurements of single particle size and composition for particulate pollution. The data are being used for single particle characterization, particle classification, source attribution, and correlating/combining with measurements from collocated instruments.

Approximately 236,000 single particle mass spectra were collected throughout the duration of the Pittsburgh Supersite experiment using RSMS-3, a third generation single particle mass spectrometer. The instrument was operated semi-continuously for 306 days, sampling particles with aerodynamic diameters in the range of 30 – 1100 nm and collecting both positive and negative ion spectra, particle size and time of detection for each particle measured. The entire data set has been fully processed and analyzed. Spectra have been clustered into 20 distinct particle classes based on the distribution of their positive ion mass peaks. Negative ion spectra were classified independently within each positive ion class. Frequency of occurrence versus particle size, month of the year and wind direction has also been calculated for the full data set, as well as within each class. Results indicate a rich array of multi-component ultrafine particles composed primarily of carbon and ammonium nitrate. Approximately 54% of all the particles measured fell into the carbonaceous ammonium nitrate (CAN) class. These particles were observed in all size bins and from most wind directions for the entirety of this study. Ubiquitous sources throughout the area, including vehicular emissions and secondary organic aerosol formation, are considered to be responsible for a larger fraction of these particles. In terms of particle number, metal containing aerosol dominated the remainder of the particle classes identified. These particles were rich in K⁺, Na⁺, Fe⁺, Pb⁺, and to

a lesser extent, Ga⁺ and Zn⁺. They tended to be smaller in size and were highly correlated with specific wind directions, facilitating the isolation of specific sources. (Bein et al. 2005)

4. Development and Evaluation of Measurement Methods

The University of Maryland has developed multi-element simultaneous GFAAS methods for determining 11 metals in three analytical groups (Group 1, predominately crustal elements, Al, Cu, Fe, Mn, and Cr; Group 2, tracers of coal and oil combustion as well as other anthropogenic sources, Se, As, Pb, and Ni; and Group 3, tracers of municipal incinerator aerosol - Zn and Cd), in ambient slurry samples collected at 30 minute intervals with the University of Maryland - Semicontinuous Elements in Aerosol Sampler, SEAS II. Addition of acid (0.2% v/v nitric acid) and ultrasonic treatment (15 min) was used to improve slurry stability and matrix composition. Wide calibration ranges were needed for analysis of the slurry samples. Improved calibration ranges were observed by using a mini carrier gas flow during the atomization stage. Compromise in analytical sensitivity varied as atomizer conditions for metals changed. Palladium (4 ug) and hydrogen (5% in Argon) were found to be effective modifiers for group one and two elements. A fast furnace temperature program was developed for Group 3. Zinc showed two-fold higher linearity in the method as compared with standard methods. Detection limits by the SEAS-II-GFAA approach were compared with traditional filter XRF, LA-ICPMS, and INAA techniques used in air quality studies. The efficacy of the analytical methods developed were applied to urban PM_{2.5} collected in Baltimore for use in a NIST fine-particulate Standard Reference material. Results obtained by the GFAA methods agreed well with the results obtained by NIST using Instrumental Neutron Activation Analysis. (Pancras 2005)

The University of California Davis coupled the Versatile Aerosol Concentration Enrichment System (VACES) to a single particle mass spectrometer (RSMS-3) to determine if the pre-concentration of particles induced by the VACES increases the hit rate of RSMS-3, as well as to monitor whether the VACES introduces any changes in the composition of the particles sampled. The goal was to increase the particle hit rate, and thus detection efficiency, of RSMS-3 in order to broaden its applicability to conditions other than polluted urban areas, for example cleaner rural sites or the stratosphere.

Results from the field evaluation indicate that VACES increased the RSMS-3 hit rate by 5-20 times at particle sizes ranging from 40 to 640 nm. VACES only enhances the hit rate by about a factor of 2 for large particle sizes because the RSMS-3 flow rates for these particles did not match the optimum operating condition of VACES. During the 3 days of measurements most of the particles were a mixture of carbonaceous material and ammonium nitrate with a variation across the spectrum from particles that were mostly carbonaceous to particles that were mostly ammonium nitrate. Both ambient and concentrated carbonaceous and ammonium nitrate composition distributions were indistinguishable with the RSMS-3, suggesting that VACES introduces an insignificant artifact for those particles. (Zhao et al. 2005)

3. Effects of dilution on mercury speciation in coal-fired power plant plumes

Dilution sampling was carried to investigate the changes in partitioning of mercury in plume of coal-fired power plants. Measurements were made on a 817 MW unit firing a blend of 60% powder river basin sub-bituminous coal and 40% mid-sulfur eastern bituminous coal. The boiler is wall-fired, with low-NO_x burners. The boiler has an ESP with SO₃ and NH₄ conditioning. An SCR is also used for NO_x control. Measurements were made over a period of 3 days at the outlet of the SCR and upstream of the air heater. Measurements were not made downstream of the SCR because of physical limitations imposed by the plant layout. Both diluted and un-diluted measurements were made at this location with a common sampling probe. A schematic of the plant with the sampling location is shown in Figure 1.

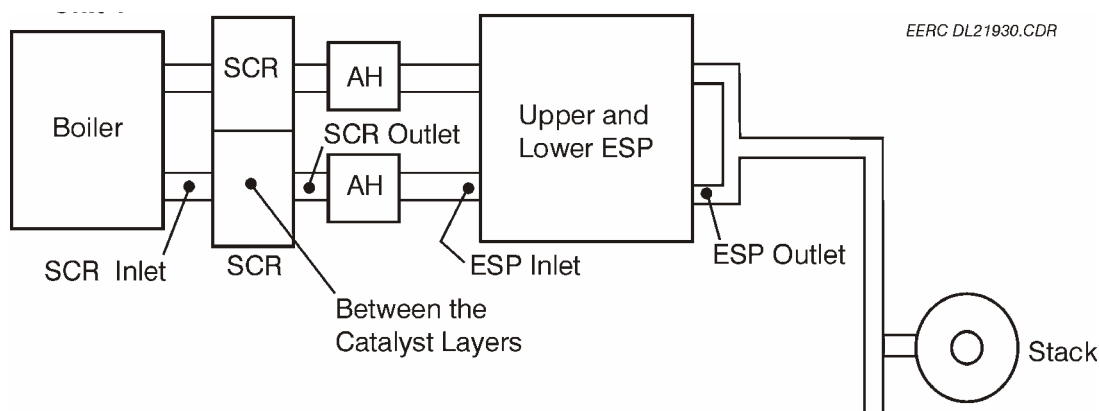


Figure 1. Schematic of power plant. Mercury sampling with the dilution sampler was performed at the location labeled SCR Outlet.

A dynamic dilution sampler was used to investigate changes in partitioning of mercury upon dilution (Figure 2). The system is designed to simulate atmospheric dilution and is constructed out of stainless steel. For the mercury testing all of the wetted components in contact with sample were coated with a 1200 Å thick layer of Silcosteel, applied by Restek Performance Coatings. In addition to the main dilution tunnel, a residence time chamber (RTC, not shown in Figure 2) was brought inline during some testing periods. With the RTC inline, the overall aging time of the diluted exhaust can be varied between 2 seconds and 5 minutes.

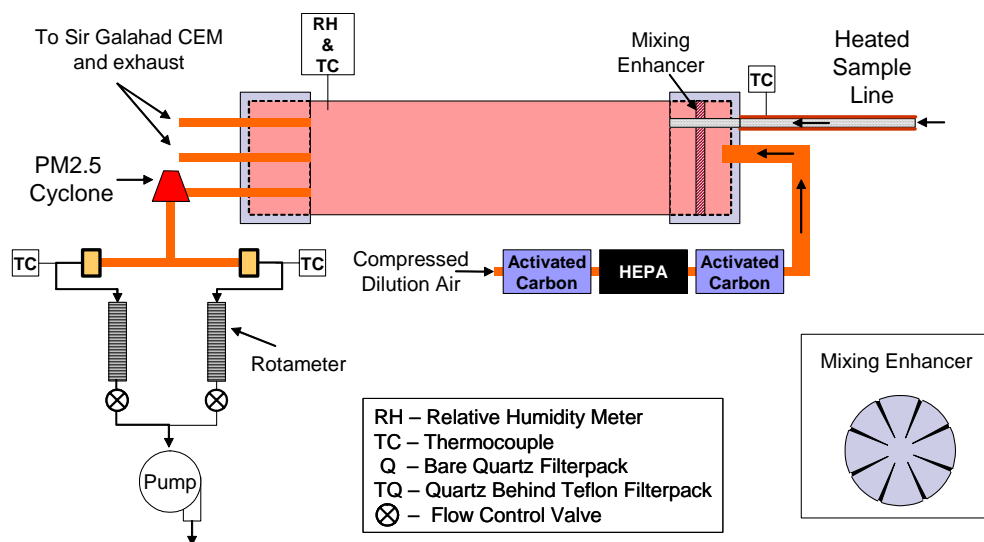


Figure 2. Schematic of CMU Dynamic Dilution Sampler (Dilution Tunnel)

Mercury sampling and speciation were via Mercury Continuous Emission Monitors (CEMs) operated by the University of North Dakota – Energy and Environmental Research Center (UND-EERC). A PS Analytical Sir Galahad CEM measured gas-phase total and elemental mercury concentrations inside the dilution sampler. These concentrations ranged from 0 to 250 ng/m³. As operated, the Sir Galahad collected sample for 5 minutes before analysis; analysis required 3 to 4 minutes and alternated between elemental and total mercury analyses. A Tekran Mercury Vapor Analyzer was used to sample the un-diluted flue gas. The Tekran measured concentrations in the range of 0 to 10 µg/m³ and alternately measured elemental mercury for 10 minutes and total mercury for 45 minutes; data points were recorded every 2 minutes in both modes.

Measurements were made over a three day period. On September 28th and on the morning of the 29th indicated significant losses of mercury inside the system. In an attempt to reduce these losses, the walls of the dilution tunnel and residence time chamber were heated to an outer surface temperature of 180 °C. Upon heating, mercury levels inside the tunnel dramatically increased presumably due to mercury desorption from the tunnel walls. This saturated the Sir Galahad analyzer, limiting data collection on the 29th. All of the experiments performed on September 30th were done with heated walls.

Blanks were measured at the beginning and end of each day of experiments by operating the system on filtered dilution air. Data collected from the dilution tunnel were blank corrected using a best estimate tunnel blank for the specific time period. The data are also dilution corrected using a period-average dilution ratio calculated using the background-corrected flue gas and dilution tunnel CO₂ concentrations. In subsequent discussion, the abbreviation DT refers to experiments performed using only the dilution tunnel, while DT + RTC refers to experiments using both the dilution tunnel and residence time chamber were online. The residence time in the two cases was approximately 2 seconds and 5 minutes, respectively.

Time series of the uncorrected (no blank or dilution correction) mercury measurements made on September 30 is shown in Figure 3. Data points in the top portion of the figure are the flue-gas mercury concentrations, which show a modest increase in total mercury concentrations over the course of the day.

The data in the lower portion of Figure 3 are mercury concentrations measured in the dilution tunnel, with different testing periods annotated. There are large changes in mercury concentrations inside the dilution sampler corresponding to changes in sampler operating conditions. For example, switching the mercury monitor from the end of the dilution tunnel to the residence time chamber at ~ 15:30 caused a large increase in measured concentrations even though a constant dilution ratio of 30 was maintained throughout this period. Figure 3 shows that this change is due in part to increasing flue gas mercury levels; note the jump in total mercury levels in dilution sampler immediately before the switch. This increase may also be due in part to desorption of mercury from the walls of the RTC, as discussed below. The dilution ratio was increased to 100 at 17:00 after which there is a slow decay in measured total and elemental mercury concentrations inside this system. This decay does show a flattening trend, but sampling was stopped at approximately 20:00, when the exhaust sampling probe was detached from the dilution system and it was operated with filtered dilution air. At the end of the day, the blank level of elemental mercury is significantly higher than at the beginning of the day. Figure 3 shows the blank slowly decaying as the system is continually flushed with filtered dilution air overnight.

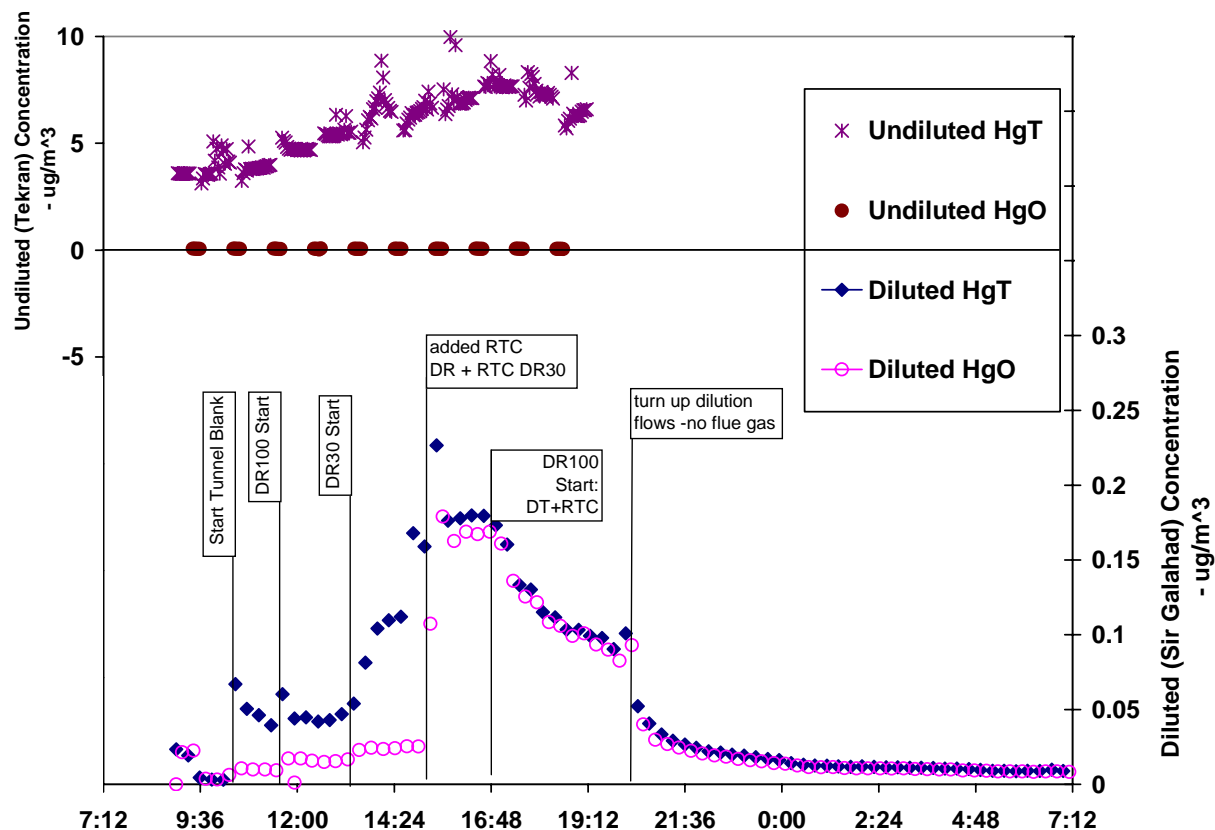


Figure 3. Time series of diluted and undiluted mercury concentration data taken on September 30

A summary of the data from each test condition is shown in Figure 4. Undiluted concentrations are an average of values from the Tekran CEM over the period when the tunnel was operated at a constant dilution ratio. The in-stack data indicate a clear predominance of oxidized mercury at this location in the plant. The total mercury concentration in the stack varied between 3 and $7.2 \mu\text{g}/\text{m}^3$. Elemental mercury ranged between 0.06 to $0.25 \mu\text{g}/\text{m}^3$. Oxidized mercury levels varied between 2.8 and $7.1 \mu\text{g}/\text{m}^3$ contributing over 90%, and generally closer to 98%, of the total mercury.

Data from the dilution sampler are shown dilution- and blank-corrected along with the flue-gas concentrations in Figure 4. It is clear that mass closure was very poor during the periods when the tunnel was unheated (on September 28 and 29). The mass balance improved somewhat when the heat-tape was added to the system (September 30). All samples except the first DT DR100 show a significant increase in dilution-corrected elemental mercury concentration relative to the flue-gas concentrations. This is a clear indicating that there is conversion of oxidized mercury to elemental mercury occurring inside the dilution sampler. The measurements made at the end of the residence time chamber show a large increase in elemental mercury and approach mass closure with the flue-gas samples.

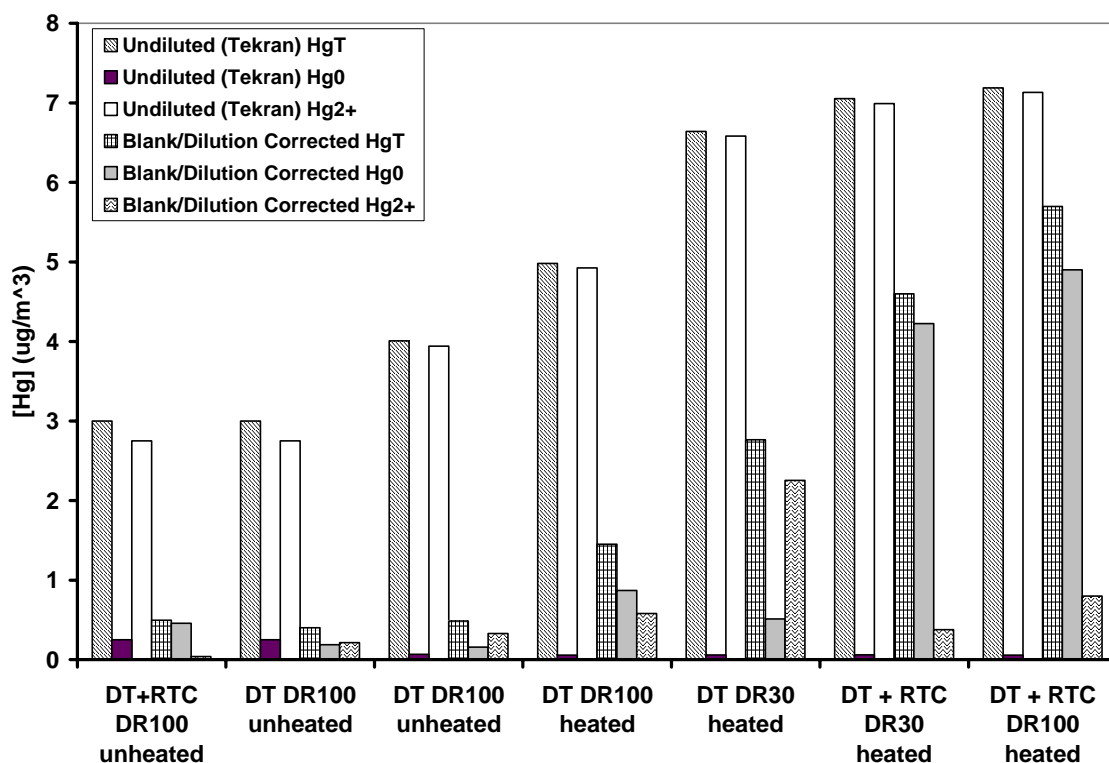


Figure 4. Flue gas and diluted sample mercury concentration. Note - Diluted samples have been corrected for tunnel blank and dilution ratio.

Mass closure and mercury transformation can be seen more clearly in Figure 5, which plots the ratios of the diluted to undiluted total, elemental and oxidized mercury concentrations presented in Figure 4. Perfect mass balance closure is a ratio of 1. The three unheated data points show very poor mass closure, with a nearly complete loss of oxidized mercury in the dilution tunnel. During these experiments, on average, only 14% of the total mercury mass measured in the flue-gas is captured by measurements in the dilution tunnel. On the other hand, the dilution corrected elemental mercury concentrations measured inside the tunnel are higher by approximately a factor of two relative to that measured in the flue gas, indicating some reduction of elemental mercury in the dilution tunnel.

Heating the tunnel walls improved mass closure. The two experiments with only the heated dilution tunnel inline show an improving total mercury mass closure relative to the flue-gas measurements; 29% of mass was recovered at DR = 100 and 42% at DR = 30. There was also a significant increase in elemental mercury levels in the diluted exhaust; elemental mercury levels in the diluted gases were on the order of 10 times those in the un-diluted gas. These two experiments also indicate that the amount of reduction occurring in the dilution tunnel increases at the higher dilution ratio. The diluted measurements with DR = 30 showed 9 times more elemental mercury than the undiluted gas, while those at DR = 100 show 16 times more.

Improved mass closure was observed in measurements made at the end of the residence time chamber. This chamber added approximately 5 minutes aging time to the system before the mercury measurements. The inclusion of the residence time chamber improved total mercury mass closure significantly, to 65% and 79% at dilution ratios of 30 and 100, respectively. However, there is a concern about whether appropriate blank corrections were applied. As indicated in the time series in Figure 3, the tunnel blank levels of total and especially elemental mercury increased significantly during the course of the day. This suggests that variable adsorption and desorption of mercury is occurring and thus complicates blank-correction of dilution tunnel measurements. These experiments showed that nearly all the mercury at the end of the RTC was in elemental form, with dilution corrected elemental mercury concentrations being a factor of 70 to 85 higher than those under non-diluted conditions. Also shown is an almost total reduction of oxidized mercury within the dilution tunnel, with 5 and 11% of the initial concentrations of flue gas oxidized mercury found in the diluted sample. As in the experiments without the residence time chamber, a larger reduction effect is seen in the run at a higher dilution ratio, suggesting a connection between dilution ratio and the mercury transformation.

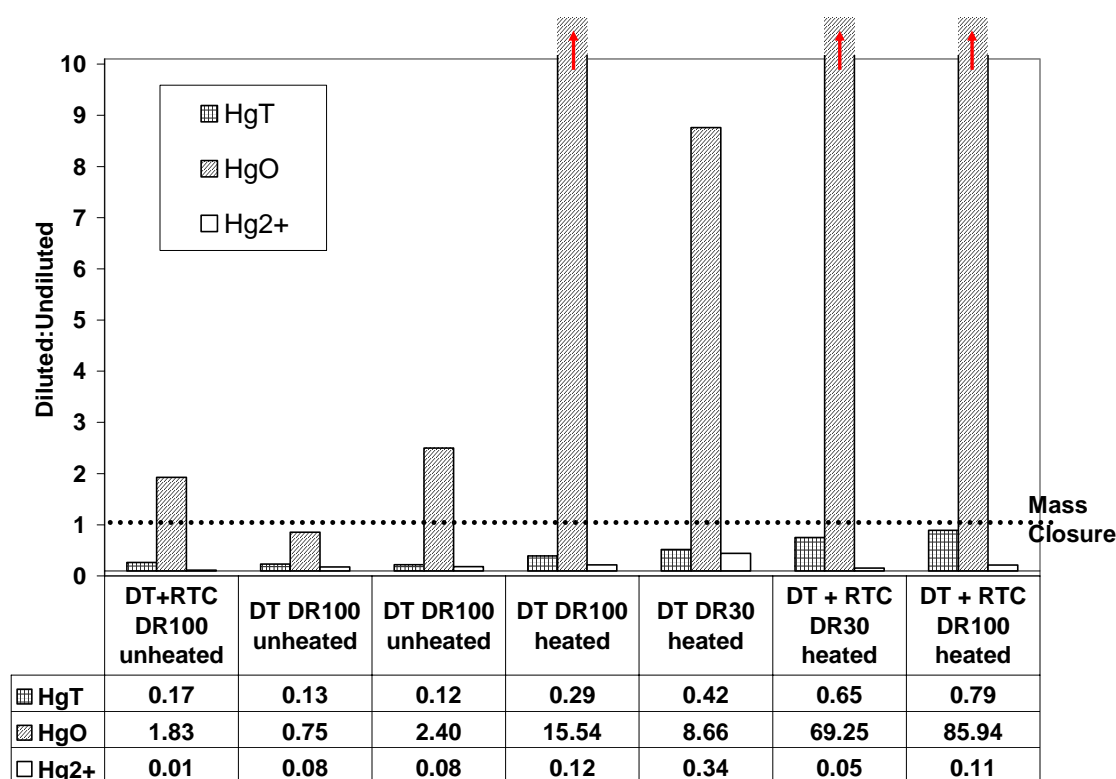


Figure 5. Ratios of diluted versus undiluted mercury concentrations. Diluted samples were blank and dilution corrected.

Although the data clearly show that oxidized mercury is being converted into an elemental state inside the dilution sampler, it is difficult to conclude whether or not these transformations are occurring on the tunnel walls or in the gas phase. However, there are

some trends that are consistent across all tests. First, there are significant losses of oxidized mercury to the tunnel walls, especially when they are not heated. The question then becomes whether the higher wall temperature reduces the absorption of mercury to the walls, or simply increasing the rate at which absorbed mercury desorbs. The fact that the mass closure improved over the course of the September 30th seems to suggest that the tunnel wall becomes 'saturated' with mercury, approaching an equilibrium state. However, the blanks of total mercury at the beginning and end of the testing on September 30th were approximately the same, while the elemental mercury blank was 4 times higher at the end of the day. This indicates that the tunnel background oxidized mercury level decreased over the course of the day and suggests that mercury was being released from the walls in elemental form during testing. If so, then some fraction of the observed transformation of oxidized to elemental mercury is occurring on the walls. Two distinct hypotheses emerge to viably explain the reduction of mercury in the tunnel:

1. Significant portions of the oxidized mercury in the flue gas are being reduced to its elemental form in the dilution tunnel in the gas phase. The magnitude of this effect increases with increasing dilution ratio and residence time – given sufficient time in the tunnel the vast majority of oxidized mercury is reduced.
2. The reduction of oxidized mercury is occurring on the heated dilution tunnel walls. Oxidized mercury is adsorbing onto the walls and surface reactions leads to its reduction to elemental form and subsequent release into the gas stream. Heating the walls increases the rate at which adsorbed mercury is released to the gas stream and influences the measurements of elemental mercury made in the system.

When the heat-tape was initially applied and mercury levels in the tunnel initially spiked (but before the analyzer saturated), the total mercury level peaked much more quickly (50 minutes earlier) than the elemental level. The implication is that the thermally-driven desorption was largely out-gassing oxidized mercury into the dilution tunnel. Since the CEM signal was saturated, nothing can be said about the relative levels of elemental and total mercury. However, this behavior suggests that reduction is not strictly occurring on the tunnel walls and lends some support to the first hypothesis.

Arriving at firm conclusions based on this data is complicated by the lack of a complete tunnel characterization during this short test period. Tunnel background mercury levels before and during tests are difficult to quantify because only two tunnel blanks -at the beginning and end of testing- were collected during the major day of data collection. In this analysis blanks from the beginning of the day was applied to data taken without the RTC online, while the blank from the end of the day was applied to the last two tests with the RTC installed. The blank levels in the tunnel at intermediate points are unknown. Attempts at interpreting the data are further complicated by the fact that our only view of the transient response of the tunnel to external heating was truncated due to instrument issues.

In future experiments, tunnel blanks taken in between test points would allow a better characterization of the transformation occurring inside the dilution tunnel. If further experiments at elevated tunnel wall temperatures are conducted, quantifying the transient response of mercury speciation in the tunnel to changes in tunnel surface temperature would be helpful. In addition, other methods for reducing wall losses might be developed. Some possibilities are applying a more inert surface coating to the tunnel, designing a tunnel that uses a sheath flow to minimize contact between the sample and the tunnel walls or running the tunnel at other surface temperatures. Development of such a sampler would be best accomplished on a laboratory or pilot-facility scale.

4. Source Apportionment of Primary Coal Emissions with SAS

RJ Lee Group continued analyzing samples collected on polycarbonate filters to determine ambient spherical aluminum silicate (SAS) particle number concentrations on both source and ambient filters. SAS particles are thought to be a unique tracer for coal-fire power plant formed from the high temperature processing of fly ash (Fisher et al. 1978; Webber et al. 1985; Eatough et al. 1996).

CCSEM methods were used to measure the size and obtain the elemental composition for 2500 individual particles from each ambient and source sample. The maximum and minimum diameter measured during the analysis was used to calculate the volume of each particle and each particle was assigned a density based on a common oxide in proportion to the elements present, determined by the EDS analysis. Images and spectra collected during the analysis were then reviewed for each individual particle to distinguish SAS from non-spherical material.

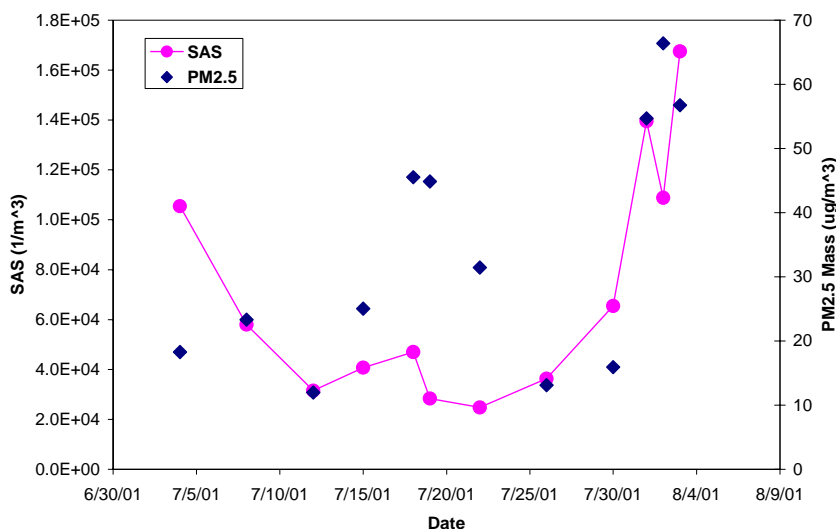


Figure 6. Time series of SAS number concentration and PM_{2.5} mass measured during the July 2001 intensive.

Time series of SAS number concentration and PM_{2.5} mass are shown in Figure 6 for the July 2001 intensive. At times the two parameters are correlated, for example

concentrations of both peaked in early August. However, in middle of July there was another spike in $PM_{2.5}$ mass concentration while SAS concentrations remain low. SAS concentrations like those of other PM parameters vary episodically presumably in response to meteorological conditions.

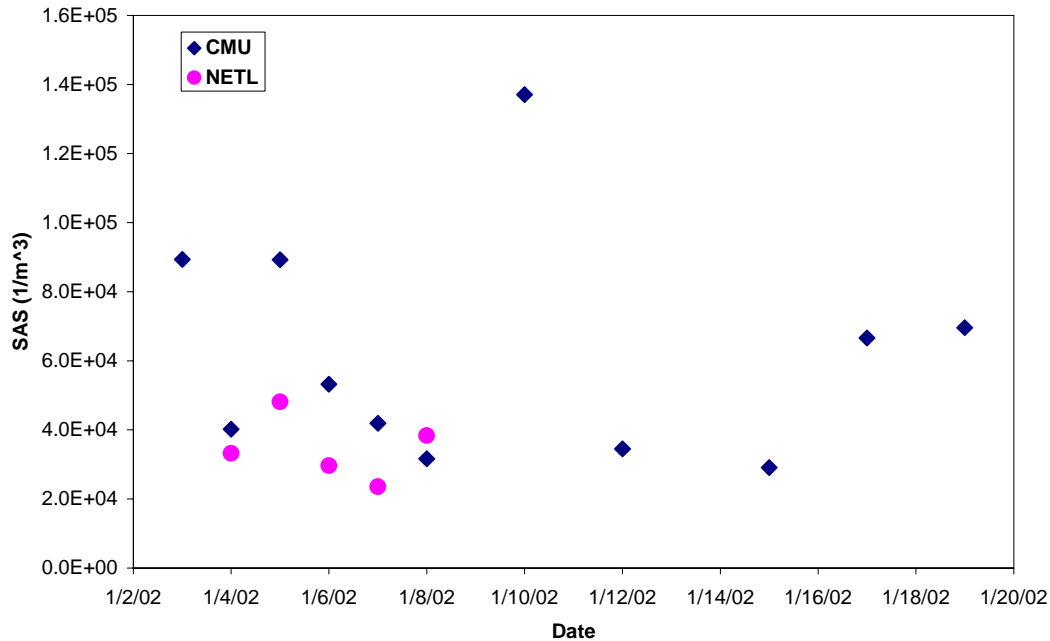


Figure 7. Time series of SAS number concentrations measured at the CMU site and the NETL site for January 2001.

Figure 7 shows time series of SAS number concentrations measured at the CMU site and the NETL site for January 2002. SAS concentrations in the winter are comparable to those in the summer. There is also reasonable in the SAS concentrations measured at the two sites on 4 of the 5 days when data are available at both sites. On the 1/5/02 SAS concentrations at the CMU site were significantly higher than at the NETL site. SO_2 concentrations at the CMU site were elevated for approximately 6 hours on this day, suggesting that a plume was impacting the site. The wind was blowing from 210° magnetic during the period with elevated SO_2 concentrations. This plume is not from the Bellefield boiler, a local coal-fired steam boiler used for district heating, which is located at a bearing 294° magnetic from the site.

The SAS number concentrations data shown in Figures 6 and 7 can be used to derive an upper bound on the contribution of primary emissions from coal-fired power plants to ambient $PM_{2.5}$ levels in Pittsburgh. The estimate is an upper bound because it assumes that coal boilers are the only source of SAS particles. To estimate the contribution of primary coal emissions to ambient $PM_{2.5}$ we need a SAS emission factor for coal combustion -- number of SAS particles/ μg primary $PM_{2.5}$ emissions. The contribution of primary coal-fired power plant emissions to the ambient $PM_{2.5}$ was determined by dividing the total number of SAS per sample collection volume by the total number of SAS per μg of coal emissions.

SAS emission factors were determined based on CCSEM analysis filters collected during source tests. Data are available from both pilot-scale and full-scale testing. Samples were collected with the Carnegie Mellon University dilution sampling system. Pilot scale measurements were made using the Combustion and Environmental Research Facility (CERF) at the Department of Energy National Energy Technology Laboratory. The CERF is a pulverized-coal combustor designed to simulate the time-temperature history of a commercial coal boiler. At full load it consumes 20 kg of pulverized coal per hour, roughly 150 kW when burning a typical US bituminous coal. Coal is injected through a swirl-stabilized burner at the top of a 3-m tall and 45-cm diameter refractory lined combustion zone. Combustion products then flow into a horizontal convective section, through two flue gas coolers, heat-traced piping, and into a bag house. The filter samples considered here were collected after the bag house. Measurements at pilot-scale were made while firing a range of common utility and industrial fuels representing: Prater Creek Coal is an eastern bituminous coal with low sulfur and ash content; Black Thunder Coal is a low-sulfur, high-calcium sub-bituminous coal from the Powder River Basin; and Bailey Mine Coal is a high-sulfur, Pittsburgh seam bituminous coal.

The full-scale measurements were made on large, modern pulverized coal boilers burning blends of eastern bituminous and western sub-bituminous coals. On boiler #1 the measurements were made upstream of the particle control device and on boiler #2 they were made downstream of the particle control device.

SAS emission factors are summarized in Figure 8 and Table 1. The emission factors vary by a factor of 7 with the emission factor for Pittsburgh #8 coal being the lowest (smallest number of SAS per $PM_{2.5}$ mass) and highest for the bituminous/sub-bituminous blend fired in Boiler #2. The measurements made at pilot-scale are comparable to those made at full scale. The most appropriate comparison is between the pilot-scale measurements made while firing Powder River Basin coal (PRB, a western sub-bituminous coal) and the full-scale data since the commercial boilers were firing a blend dominated by western sub-bituminous coal. The emission factor measured while firing PRB at pilot-scale falls in between the results from the full-scale tests. Another factor is the potential effects of particle control technology on the SAS emission rate. A bag house is used for particle control on the pilot-scale unit while measurements at full scale were made both before (Boiler #1) and after (Boiler #2) an ESP. The agreement between the pilot-scale data and two full-scale tests suggests SAS emission rate as a fraction of $PM_{2.5}$ mass is not sensitive to particle control technology. The data do suggest some rank dependence in the emission rate of SAS with the SAS emissions of the lower rank sub-bituminous coals being higher than that of the higher rank eastern bituminous coals.

Table 1. SAS emission factors (number of SAS particles/ μg primary $\text{PM}_{2.5}$ emissions) for different fuels.

Test	SAS/ μg
<u>Pilot-Scale Combustor</u>	
Prater Creek Coal (Test 1)	2.16E+05
Prater Creek Coal (Test 2)	2.14E+05
Pittsburgh #8 Coal	1.25E+05
Powder River Basin	5.81E+05
Prater Creek Coal-Wood Blend	3.16E+05
<u>Full-Scale Boiler</u>	
Boiler #1	4.40E+05
Boiler #2	7.11E+05

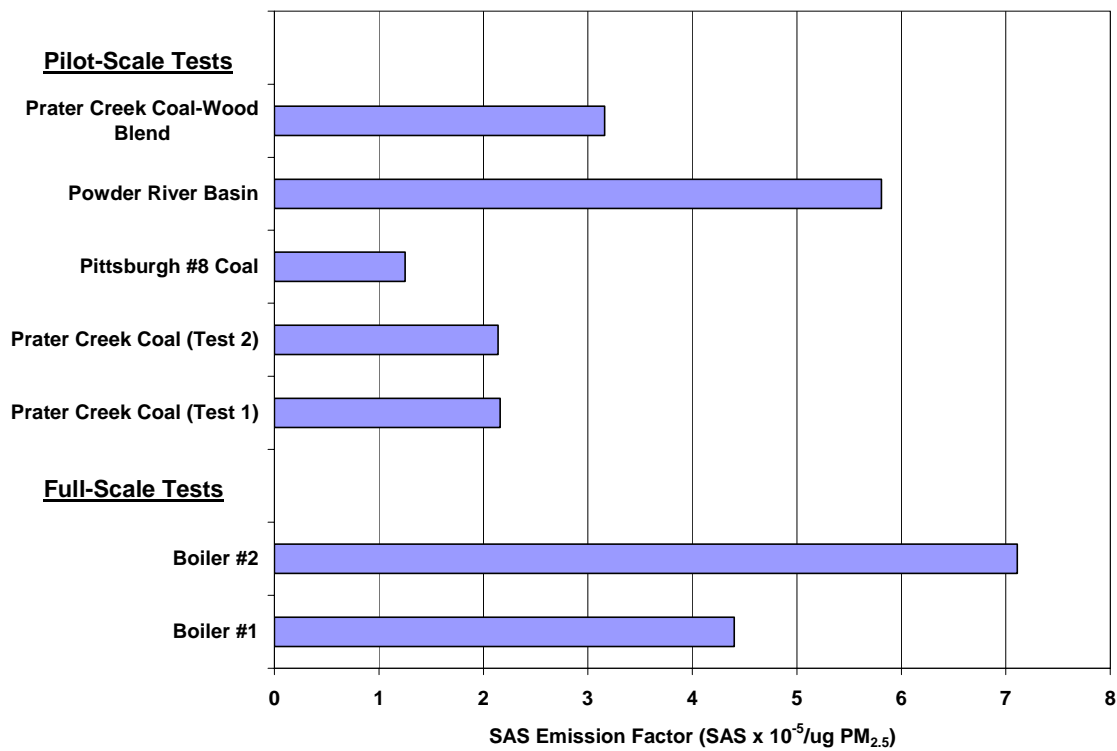


Figure 8. SAS emission factors as a fraction of $\text{PM}_{2.5}$ mass measured during pilot-scale and full-scale boiler testing.

Time series of estimated $\text{PM}_{2.5}$ mass contributed by primary emissions from coal combustion for the Summer 2001 Intensive are shown in Figure 9. Using the Pittsburgh #8 coal emission factor produces the highest estimate while the Boiler #2 profile produces the lowest estimate. Using these two profiles the monthly-average coal contribution to $\text{PM}_{2.5}$ mass for July 2001 is between $0.09 \mu\text{g}/\text{m}^3$ (boiler #2 profile) to 0.55

$\mu\text{g}/\text{m}^3$ (Pittsburgh #8 coal) -- 0.25% to 1.6% of the ambient monthly average $\text{PM}_{2.5}$ mass. The monthly average contributions for January 2002 are essentially the same on an absolute basis, 0.08 to 0.58 $\mu\text{g}/\text{m}^3$ of $\text{PM}_{2.5}$ mass (0.5% to 3.1% of ambient $\text{PM}_{2.5}$ mass). On a relative basis, primary coal emissions contribute a larger fraction of the $\text{PM}_{2.5}$ mass in the winter than the summer because ambient $\text{PM}_{2.5}$ mass is lower in the wintertime. The agreement in the absolute contribution of primary coal in the summer and winter indicate that the CMU is site is not being significantly impacted by seasonal emissions from local boilers used for heating purposes. These estimates are consistent with expectations that primary coal emissions are a small contributor to ambient fine particulate matter mass.

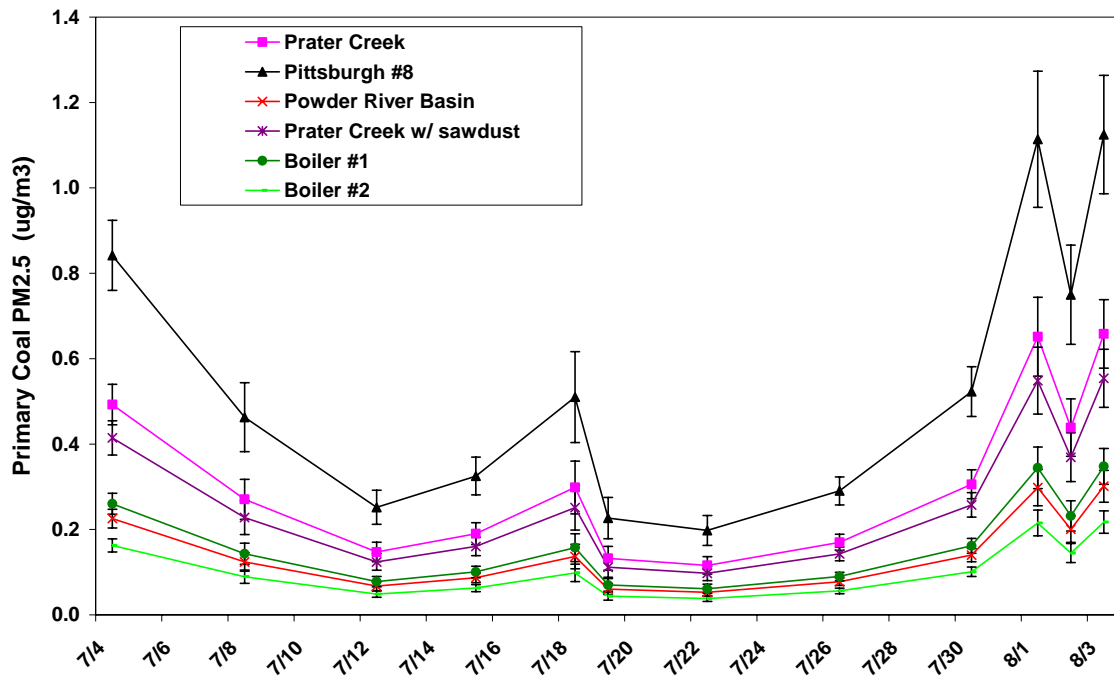


Figure 9. Estimate $\text{PM}_{2.5}$ mass contributed by primary coal emissions to the Pittsburgh Supersite during the Summer 2001 intensive.

5. Source Apportionment using PMF and UNMIX

An objective of the Pittsburgh Air Quality Study was to determine the major sources of $\text{PM}_{2.5}$ in the Pittsburgh region. Daily 24-hour averaged filter-based data were collected for 13 months, starting in July 2001, including sulfate and nitrate data from IC analysis, trace element data from ICP-MS analysis, and organic and elemental carbon from the thermal optical transmittance (TOT) method and the NIOSH thermal evolution protocol. These data were used in two source-receptor models, Unmix and PMF.

The Unmix model created a six-factor solution with the source compositions shown in Figure 10 and source contributions shown in Figure 11. Some combinations of the species considered resulted in a solution that was not feasible. The combination of species that provided the best solution included Ca, Ti, Cr, Mn, Fe, Zn, Mo, Cd, sulfate,

nitrate, OC, and EC. Three parameters designed to evaluate the model results are the minimum R^2 , the signal-to-noise ratio, and the strength. The R^2 value is related to the proportion of variance of each species explained by the factors. For all species, the minimum R^2 value is recommended to be greater than 0.8. For the selected species, the minimum R^2 value was 0.86. The minimum signal-to-noise ratio is the smallest estimated signal-to-noise ratio for any of the factors in the model, recommended to be greater than 2. A value of 2.32 was obtained using this dataset. The strength is a measure of the confidence in the model. Strength is recommended to be greater than 3, but with some datasets this is unachievable and thus a strength less than 3 may still be acceptable (Henry, personal communication). For this dataset, it was impossible to find a combination of species yielding a strength greater than 3, and the final solution had a strength of 1.41.

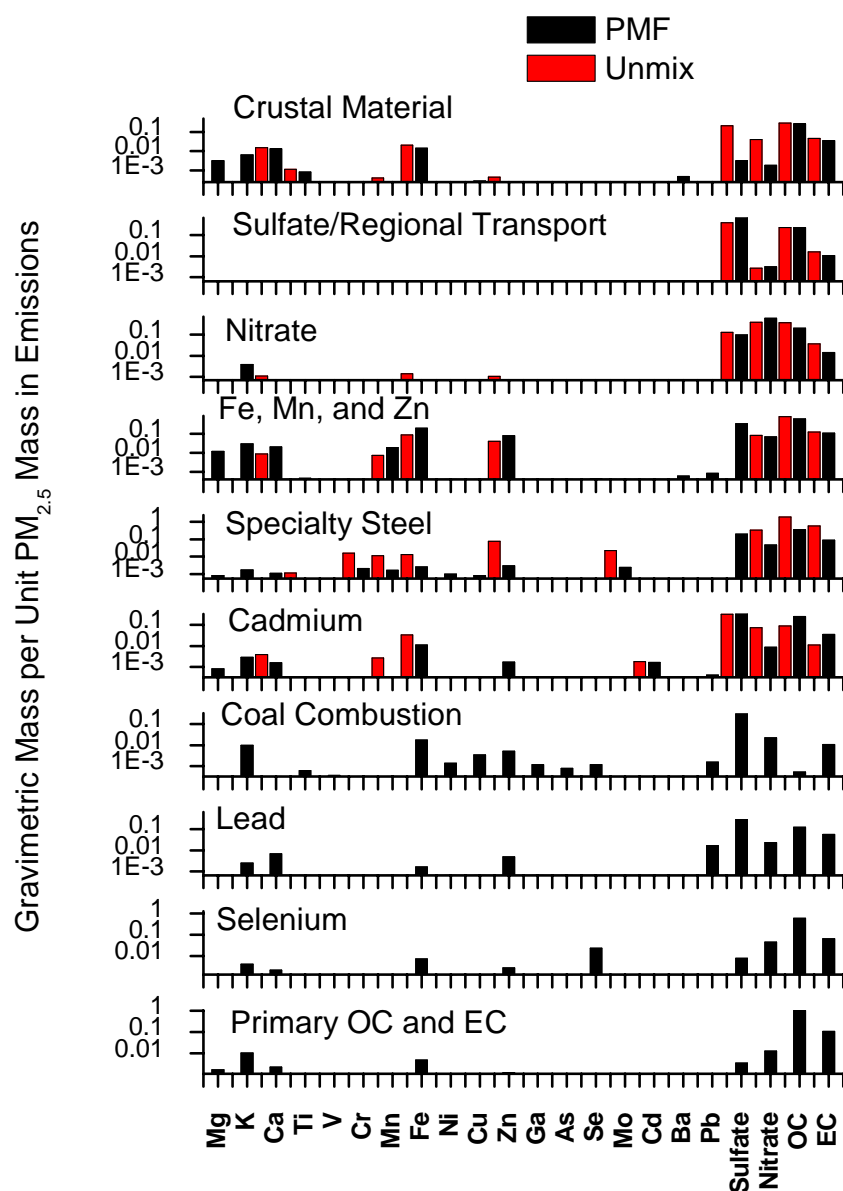


Figure 10. Unmix source compositions apportioned by $PM_{2.5}$ mass. All ten factors are outputs of PMF, while only the first six factors are outputs of Unmix. Although the Unmix regional transport factor includes primary OC and EC, it is graphed for comparison with the PMF sulfate factor.

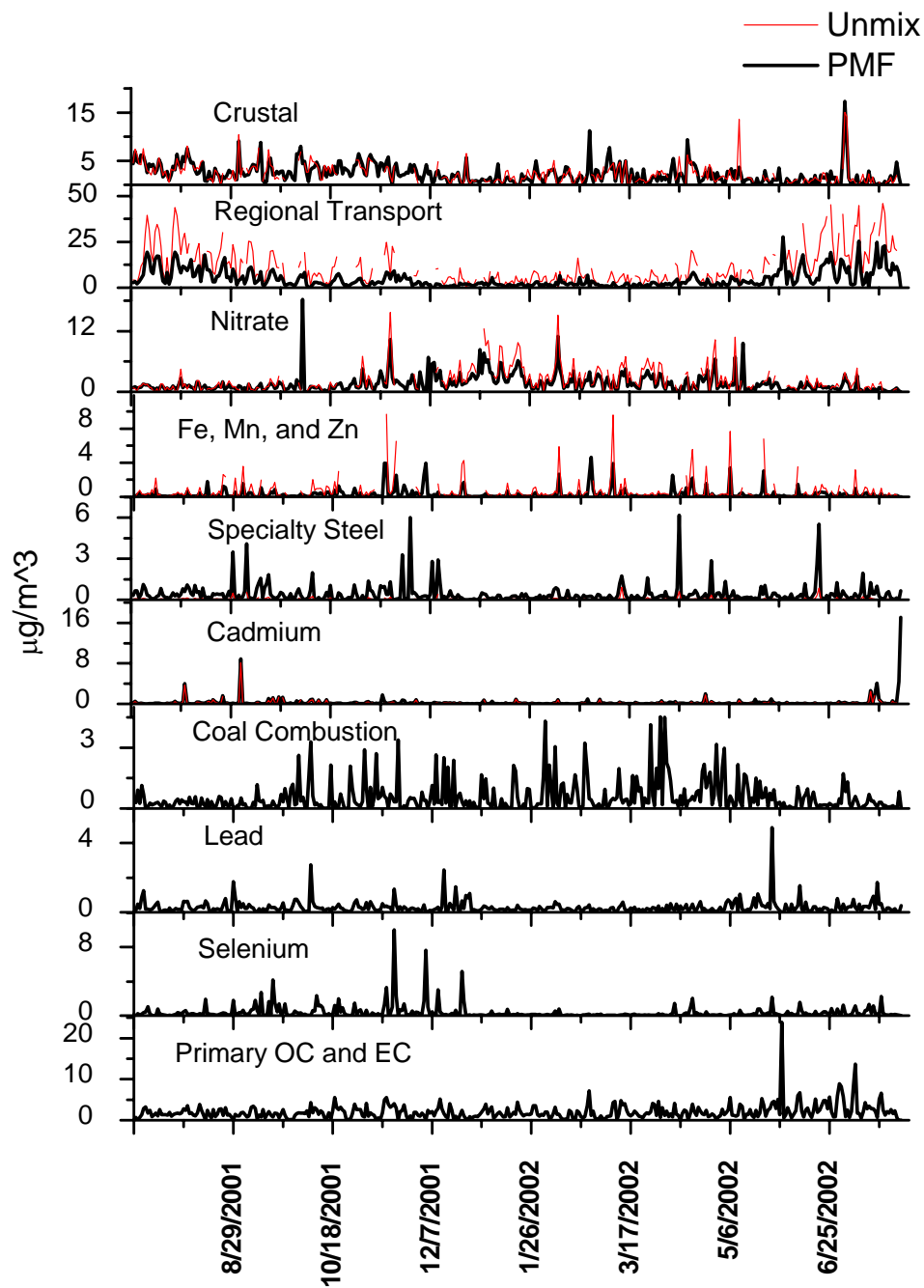


Figure 11. PMF and Unmix source contributions apportioned by PM_{2.5} mass. All ten factors are outputs of PMF, while only the first six factors are outputs of Unmix. Note that for comparison purposes, the Unmix regional transport factor is compared to the PMF sulfate plus primary OC and EC factors.

Species for which the ambient data correlated strongly with the source contributions (correlation coefficient greater than 0.7) allow determination of the source types. The six factors in the model have been designated a crustal material factor, a regional transport factor, a nitrate factor, an Fe, Mn, and Zn factor, a specialty steel production factor, and a cadmium factor. Descriptions of the nature of the factors, such as their contributions to $PM_{2.5}$ mass on a seasonal basis as well as a yearly average, are described below. To determine the mass contribution to $PM_{2.5}$, the total $PM_{2.5}$ mass was included as a species in the model, and the calculated source compositions and contributions were normalized by the Unmix-apportioned $PM_{2.5}$ mass. Because $PM_{2.5}$ is used as a fitting species, together these six factors account for all of the $PM_{2.5}$ mass. Figure 12 shows the Unmix source contributions normalized by the Unmix-apportioned $PM_{2.5}$ mass, averaged monthly and for the entire study.

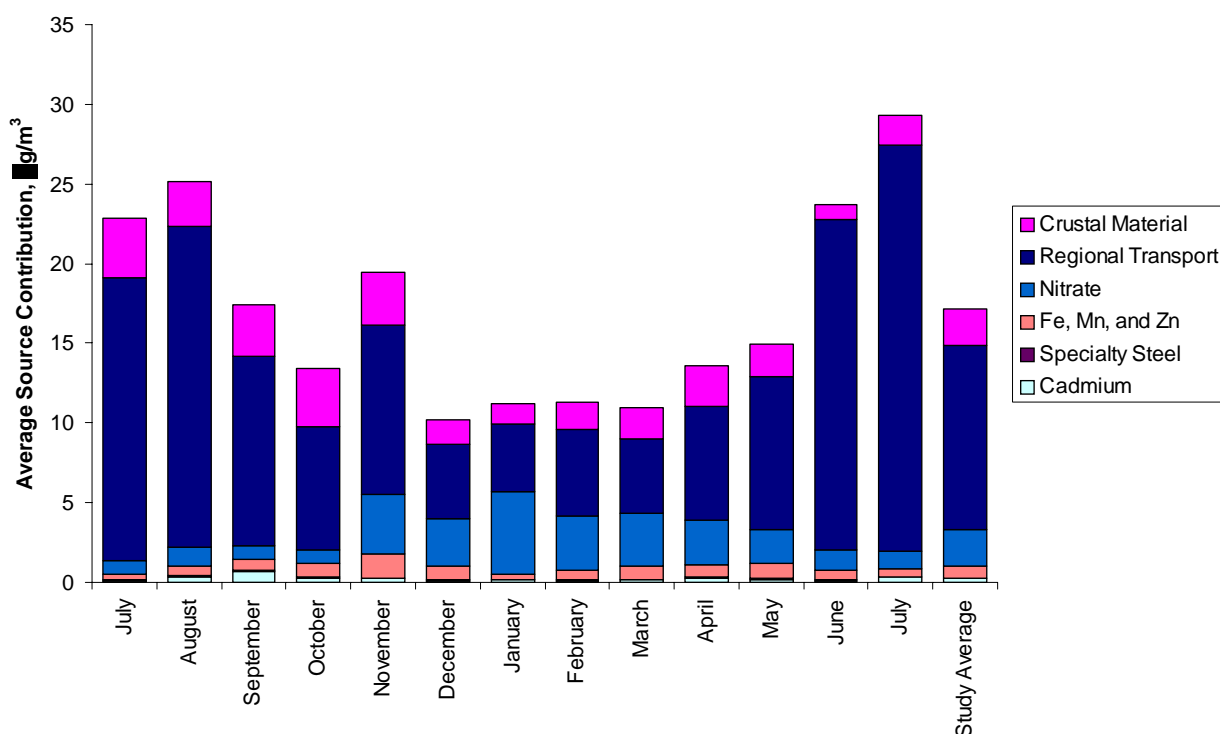


Figure 12. Monthly average Unmix source contributions. Height of the bars corresponds to the monthly average $PM_{2.5}$ mass measured with a TEOM. The study average represents the average source contributions from July 11, 2001 through July 31, 2002. Unmix uses $PM_{2.5}$ mass as a fitting species so the mass of $PM_{2.5}$ unexplained by Unmix is less than 1%

PMF allows inclusion of more species in the model due to the consideration of uncertainties that enables handling of missing and below detection limit data. Species included in the PMF solution are $PM_{2.5}$ sulfate, nitrate, OC, EC, Mg, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ga, As, Se, Mo, Cd, Ba, and Pb. We can identify a tracer species for each factor based on the source compositions shown in Figure 10. However, a better indication is the correlation of the species ambient concentration with the PMF-modeled

source contribution. A correlation greater than 0.7 is a good indication of a tracer species. Based on the tracer species for each factor, the factors were defined as crustal material (Ca and Ti tracers), sulfate, nitrate, an Fe, Zn, and Mn factor, specialty steel (Mo and Cr tracers), cadmium, coal combustion (Ga tracer), lead, selenium, and primary OC and EC (OC and EC tracers).

A comparison of compositions and contributions for factors found by both Unmix and PMF are shown in Figures 10 and 11. These source categories include crustal material, sulfate, nitrate, steel production, specialty steel, and cadmium. The factors not found by Unmix but found by PMF are described below, and their source compositions and contributions are also shown in Figures 10 and 11. Figure 13 shows the average PMF source contributions apportioned by average $PM_{2.5}$ mass concentration. A total of 22% of the $PM_{2.5}$ measured with the TEOM is not apportioned to any source by PMF. This missing mass could be explained by species not included in the model, such as particulate ammonium, or the presence of water in the particles that was measured as $PM_{2.5}$. If all of the sulfate is assumed to be ammonium sulfate, the missing mass fraction decreases to 13%. Assuming that all of the nitrate is ammonium nitrate as well decreases the missing mass fraction to 9%.

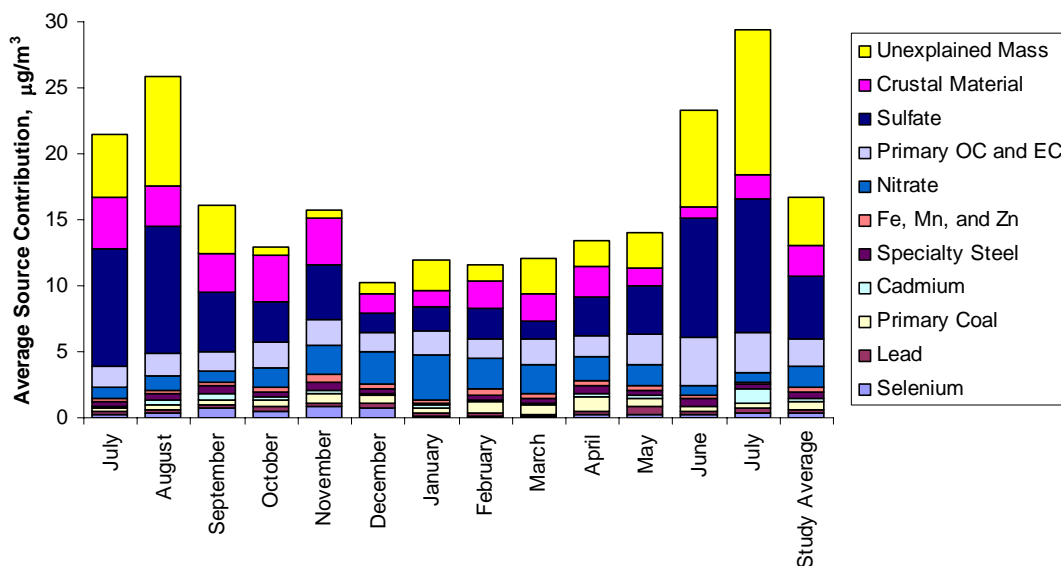


Figure 13. Monthly average PMF source contributions. Height of the bars corresponds to the monthly average $PM_{2.5}$ mass measured with a TEOM. The study average represents the average source contributions from July 11, 2001 through July 31, 2002. The unexplained mass is the difference between the monthly average $PM_{2.5}$ mass and the sum of the monthly averaged source contributions from each factor.

Agreement between the two models is generally quite good, both in composition of sources and source contribution trends. However, there are a few significant differences. While source contributions track well for the two models, showing similar trends in

concentration with time, the magnitude of the contribution does not agree for some factors. Figure 14 shows the results of a linear regression of the Unmix source contributions against the same factors found by PMF. For this comparison, the PMF sulfate and primary OC and EC factors were added together for comparison with the Unmix regional transport factor. The PMF sulfate and primary OC and EC factors were added together for comparison with the Unmix regional transport factor.

The R^2 values for all factors are reasonable and statistically significant, ranging from 0.64 for crustal material to 0.99 for the cadmium factor. The slope of the regression line, however, ranges from 0.41 for the Fe, Mn, and Zn factor to 6.2 for the specialty steel factor. The crustal material and the cadmium factors are within 20% in source contribution magnitude, suggesting that results are robust for these factors. PMF apportions more mass to the specialty steel factor due to the inclusion of 2% of the sulfate mass. PMF apportions less mass to the sulfate and primary OC and EC factors, the nitrate factor, and the Fe, Mn, and Zn factor. The apportionment of less mass to the sulfate and primary OC and EC factors by PMF as compared to the Unmix regional transport factor is likely due to Unmix fitting the model to total $PM_{2.5}$, while PMF has a significant fraction of unexplained mass. For the nitrate and Fe, Zn, and Mn factors, the difference is due to the apportionment of OC and EC. Unmix apportions 17% of the OC mass and 17% of the EC mass to the nitrate factor whereas PMF apportions 7% OC mass and 5% EC mass to the nitrate factor. For the Fe, Mn, and Zn factor, the apportionment is 12% of the OC mass and 18% of the EC mass explained by Unmix, but only 4% of the OC mass and 7% of the EC mass is explained by the same PMF factor. Results from previous comparisons of PMF and Unmix show similar conclusions: convergence for some factors but poor agreement for others (Poirot et al., 2001; Maykut et al., 2003).

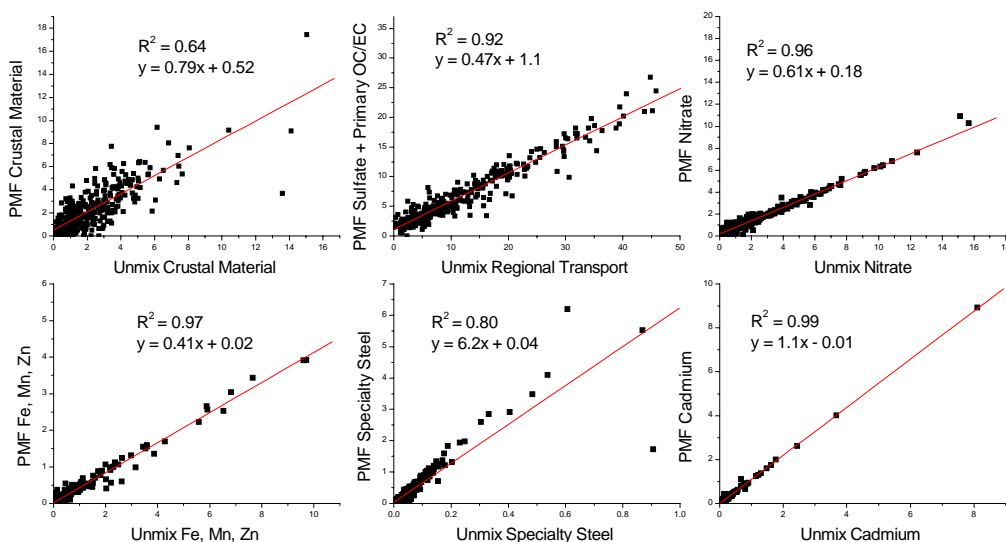


Figure 14. Linear regression results of PMF and Unmix source contributions.

In comparing Figures 12 and 13, the average source contributions as a percent of average $PM_{2.5}$ mass are within a few percent for the two models for all factors, with the

exception of the Unmix regional transport factor (68%) and the sulfate and primary OC and EC factor for PMF (total 40%). Unmix apportions all the mass, while on average PMF apportions only 78% of the mass, so some discrepancy is expected. PMF is more effective at discerning between primary and secondary OC; Unmix does not distinguish between the two and therefore can only give a large factor that is general regionally transported material and is not very informative from a policy-making perspective.

CONCLUSIONS

Significant progress was made this project period on the analysis of ambient data, source characterization, and source apportionment activities. This report highlights results from mercury emission measurements made using a dilution sampler. The measurements clearly indicate that mercury is being transformed from an oxidized to an elemental state within the dilution. Because of the effect of wall losses it is not clear whether or not this transformation occurs in the gas phase or due to some interaction with the sampler walls.

This report also presents results from an analysis that uses SAS as a marker for primary PM_{2.5} emitted from coal combustion. Primary emissions from coal combustion contribute only a small fraction of the PM_{2.5} mass at the Pittsburgh site. Ambient SAS concentrations also appear to be reasonably spatially homogeneous. Finally, SAS emission factors measured at pilot-scale appear consistent with measurements made at full-scale.

The Unmix and PMF models have been used to estimate the contribution of different sources to the PM_{2.5} mass concentrations in Pittsburgh. Comparison of the two models shows similar source composition and contribution for five factors: crustal material, nitrate, an Fe, Mn, and Zn factor, specialty steel production, and a cadmium factor. PMF found several additional factors: coal combustion, a lead factor, and a selenium factor assumed to be related to coal combustion. The PMF model found a sulfate factor separate from the OC and EC associated with primary emissions, while Unmix grouped these three species together into a single factor. Comparison between source contributions for the similar factors shows reasonable agreement between the two models. The sulfate factor shows the highest contribution to local PM_{2.5} with an annual average contribution of approximately 28% (from PMF). The nitrate, crustal material, and primary OC and EC factors also show significant contributions on the order of 10-14%. The sulfate factor is affected by photochemistry and therefore shows maximum values in summer. The nitrate factor is temperature sensitive due to the volatility of nitrate; maximum values of particulate nitrate occur in winter. The crustal material and vehicle sources somewhat more constant contributions throughout the year. The remaining factors contribute on a smaller scale and are defined by plume events, with peaks in concentration distinctly higher than average concentration.

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1. "Investigation of nucleation bursts in the Pittsburgh air quality study", 6th International Aerosol Conference, Taipei, Taiwan, September 2002 (C. O. Stanier, A. Y. Khlystov, and S. N. Pandis).
2. "Monitoring of water content of ambient aerosol during the Pittsburgh Air Quality Study" 6th International Aerosol Conference, Taipei, Taiwan, September 2002 (A. Y. Khlystov, C. O. Stanier, D. Vayenas, and S. N. Pandis).

3. Performance of the Aerodynamic Particle Sizer 3320 during the Pittsburgh Air Quality Study (PAQS)” 6th International Aerosol Conference, Taipei, Taiwan, September 2002 (A. Khlystov, C. Stanier, and S. N. Pandis).
4. “Sulfate-ammonia-nitric acid interactions in an urban area” 6th International Aerosol Conference, Taipei, Taiwan, September 2002 (S. Takahama, A. Khlystov, B. Wittig, S. V. Hering, C. Davidson, A. Robinson, and S. N. Pandis).
5. “Sampling artifacts during measurement of ambient carbonaceous aerosol” 6th International Aerosol Conference, Taipei, Taiwan, September 2002 (R. Subramanian, A. Y. Khlystov, J. C. Cabada, S. N. Pandis, and A. L. Robinson).
6. “Formation and properties of regional aerosol: Some insights from the Pittsburgh Air Quality Study”, NASA-GSFC, Greenbelt MD, May 2002,(C. Stanier, A. Khlystov, S. Rees, J. Cabada, A. Robinson, C. Davidson, and S. N. Pandis)
7. “Seasonal composition of PM_{2.5} and performance of the Federal Reference Method in Pittsburgh”, PM_{2.5} and Electric Power Generation, Pittsburgh, April 2002 (S. L. Rees, S. Takahama, A. L. Robinson, A. Khlystov, and S. N. Pandis).
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13. “Automated measurements of dry and wet ambient aerosol distributions”, Annual Meeting of AAAR, Portland, Oregon, October 2001 (A. Y. Khlystov, W. R. Chan, C. O. Stanier, M. Mandiro, and S. N. Pandis)

14. "Continuous measurements of ammonia and ammonium in ambient air", Annual Meeting of AAAR, Portland, Oregon, October 2001 (A. Khlystov, J. Sauser, R. Otjes, and S. N. Pandis).
15. The contribution of secondary organic aerosol to PM_{2.5} concentrations in Pittsburgh, AGU Fall Meeting 2002, San Francisco CA Dec. 2002 (J. C. Cabada, S. N. Pandis, A. L. Robinson, R. Subramanian, A. Polidori, and B. Turpin).
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